

PLUTONIUM RADIONUCLIDES IN THE GROUNDWATERS AT ENEWETAK ATOLL

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Abstract

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In 1974 a groundwater programme was initiated at Enewetak Atoll to study systematically the hydrology and the groundwater geochemistry on selected islands of the Atoll. The programme provides chemical and radiochemical data for assessment of water quality on those islands designated for rehabilitation. These and other data are used to interpret the mechanisms by which radionuclides are cycled in the soil-groundwater system. Because of the international concern over the long-term buildup, availability, and transport of plutonium in the environment, this programme emphasizes analysis of the element.

The results of the study show that on all islands sampled, small quantities of plutonium radionuclides have migrated through the soil columns and are redistributed throughout the groundwater reservoirs. The observed maximum surface concentrations are less than 0.02% of the maximum recommended concentration for drinking water. Concentrations of ^{137}Cs are found to correlate with water freshness, but those of $^{239,240}\text{Pu}$ show no such relationship. The mechanisms moving $^{239,240}\text{Pu}$ through the groundwater reservoirs are independent of the processes controlling the cycling of ^{137}Cs and fresh water. A reasonable linear correlation is found between mean surface-water concentrations and soil burdens. The quantities of $^{239,240}\text{Pu}$ migrating to the groundwater surface layers do not correlate with any other known differences in the physical, chemical or biological characteristics of the islands.

1. Introductions and Background

Enewetak, the northwesternmost atoll in the western (Ralik) chain of the Marshall Islands, was one of the U.S. nuclear testing sites in the Pacific. The Atoll consists of 36 islands on an elliptical coral reef encompassing a lagoon with an area of 931 km². The islands, which were given alphabetic code names during the U.S. occupancy, are identified and shown in Fig. 1. The total land area is about 7 km² and the maximum ground elevation above mean sea level is less than 4m. The islands vary in size from small sand bars of a few hundred square meters in area to about 1.3 km². The largest islands, hence the most important for future habitation, are Fred, Elmer, and David in the southeast and Janet in the north. The Atoll has a tropical marine climate characterized by **uniformity** of air pressure, temperature, cloudiness, and humidity. Wind **conditions** are somewhat variable, but moderate easterly trade winds **predominate** over the year. Precipitation is extremely variable over the **year**. Table 1 compares monthly rainfall measurements to the monthly mean **and range** for the past 13 years. It is evident that data from a single

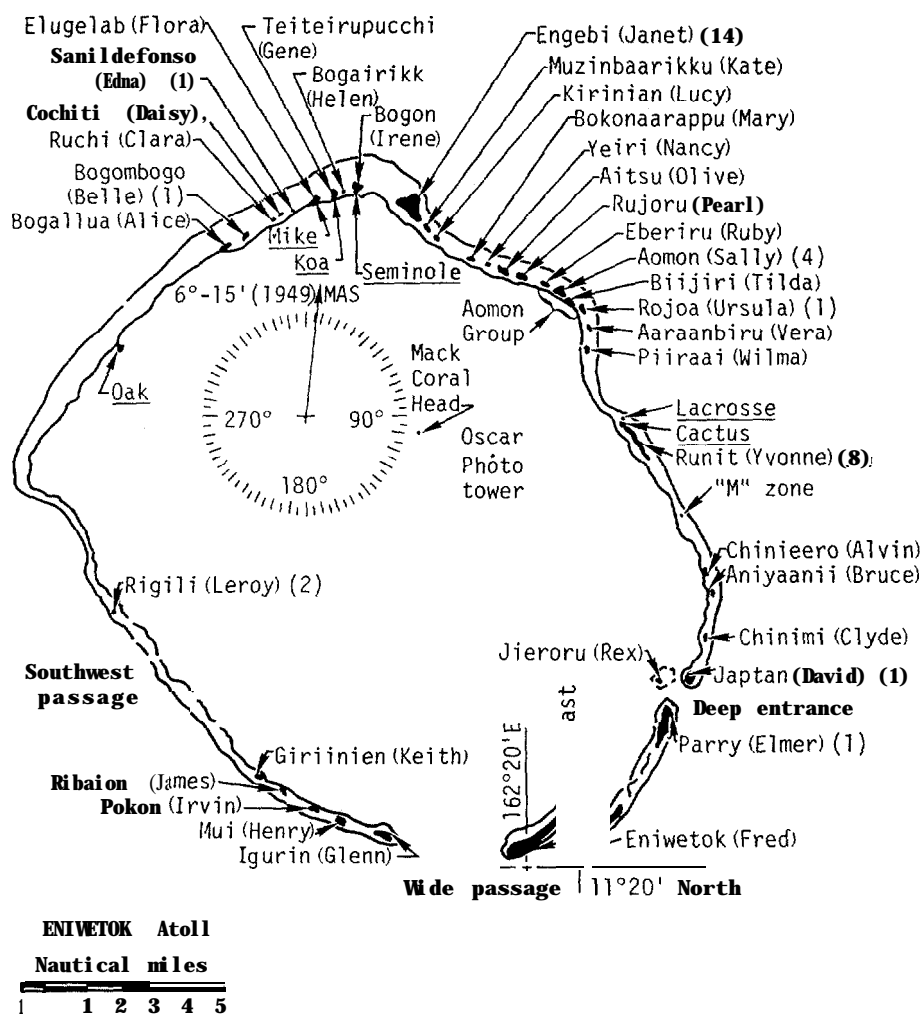


FIG. 1. The islands of Eniwetok Atoll. The names of the major craters are underlined. The number of groundwater wells is given for each island in parentheses after the island name.

TABLE I. Monthly Rainfall at Eniwetok Atoll.

	Mean 1963-1973 (cm)	Range 1963-1973 (cm)	Total 1974 (cm)
Jan	3.45	0.76- 9.55	16.3
Feb	2.31	1.02- 3.99	1.80
March	3.73	0.33-11.4	7.36
April	8.46	0.48-26.82	31.01
May	16.81	0.91-26.82	11.61
June	12.07	4.62-26.98	14.96
July	7.63	9.70-31.19	9.75
Aug	19.27	7.56-34.36	35.66
Sept	24.91	10.87-51.63	26.92
Oct	27.02	16.10-62.33	32.46
Nov	13.43	8.56-19.32	19.9
Dec	7.56	2.05-23.31	5.0

season provide no indication of the limits of monthly variation. Since groundwater recharge is directly related to rainfall, the accurate assessment of the amount of rainfall is an important part of this study. The total of 43 nuclear tests at the Atoll during the 1948-1958 testing periods resulted in the spread and deposition of nuclear debris throughout the Atoll environment. The test years during this interval were 1948, 1951, 1952, 1954, 1956, and 1958.

An extensive radiological survey of the Atoll was conducted from late 1972 through early 1973. Its purpose was to document the levels and distributions of the longer-lived radionuclides still present in the Atoll environment and to examine the major components of most significance to the radiological dose to the Marshallese people who are about to reinhabit the Atoll. The survey results were published in October 1973 [1]. It was concluded [1] that the terrestrial food chain potentially could contribute far greater doses to a returning population practicing subsistence agriculture than could the present external radiation levels, the marine food chain, or the inhalation pathways.

The models used for assessing the dose from terrestrial radioactivity did not take into account the possibility that some of the residual radionuclides might become less available with time by processes other than radioactive decay. For example, if erosion of soil material, induced by the infiltration of rainwater or groundwater in the soil column, causes solution of the bound radionuclides or migration of the particulates, loss of radionuclides through the groundwater aquifers could significantly reduce some of the existing soil burdens. The roots of breadfruit, pandanus, and other plants have been found to penetrate to the groundwater surface layer at these Pacific Islands [2]. There is little question that the groundwater partially sustains a number of phreatophyte plants on the Atoll, and radionuclides must be taken up by plants from the groundwater and hence recycled to the soil surface through litter deposition. Data collected during the survey [1] were not sufficient to evaluate the importance of these pathways.

It was assumed also that the Enewetak people, after returning to their Atoll, will continue the practice of using catchment rainwater for drinking and that underground lens water will not be a major part of their diet. Rainwater is preferred for drinking, but potable groundwater can be used for household purposes and the watering of crops (as it is presently used on Bikini Island), and during the dry season it must be relied on for drinking (as it has been at Bikini Island) when rain cisterns are completely empty. Therefore, both the chemical and radiological quality must be assessed before rational decisions can be made regarding the use and management of this water resource.

To fulfill this need, a groundwater program was initiated at Enewetak Atoll in 1974 to study systematically the hydrology and the groundwater geochemistry on selected islands of the Atoll. This project is a joint study between the Lawrence Livermore Laboratory and the University of Hawaii. From the chemical and radiochemical data being collected we expect to gain an understanding of the transport of radionuclides in the soil/groundwater/vegetation systems. With such understanding, we can proceed to assess the long-term dose to an indigenous population practicing subsistence agriculture on the contaminated and the relatively uncontaminated islands, and to assess the present and future quality of the groundwater. More extensive discussions on the principles of fresh groundwater occurrence on small oceanic islands

can be found in other publications [3, 4, 5]. The discussion of these principles and how they relate to the geography and geology of Enewetak Atoll is beyond the scope of this report.

Elevated plutonium levels (greater than deposition levels from world-wide fallout) were detected throughout the Atoll environment [1]. Several reviews [6, 7, 8] have emphasized the growing concern about man's ignorance of the biogeochemistry of this element in relation to its long-term buildup, availability, and transport in the environment. This program has emphasized measurements of plutonium for two reasons. The first is to understand better the pathway and movement of plutonium through a soil column, to and through an accessible groundwater reservoir, on an atoll contaminated with close-in fallout debris. The second is to establish a data base for the assessment of water quality. Although ^{60}Co , ^{90}Sr , ^{137}Cs , ^3H , and ^{125}Sb as well as plutonium have been detected in some but not necessarily all groundwater samples, this report discusses only our findings for plutonium; other data are presented, however, when they are pertinent to the interpretation of our plutonium results.

2. Experimental Methods

Arrangements were made early in 1974 with project personnel of USAF EXPOE (Exploratory Program on Enewetak) to drill 33 test wells and to case each with slotted PVC casing. The wells are located on nine different islands. Groundwater was encountered at all sites at depths of 2.5 m to 3 m below ground surface. The number and locations of the wells were chosen after consideration of both scientific and practical factors. Information was desired on the hydrology, the quantity, and the chemical and radiological composition of fresh and brackish water on various islands as a function of island area, shape, and position on the Atoll. Radiological and chemical data were desired on those islands designated for rehabilitation, but also on other islands to gain an understanding of the processes controlling the distributions of radionuclides on islands with different radiological burdens and amounts of vegetation. More mundane factors such as accessibility of the island to drilling equipment, cost, transportation, scheduling, and weather had to be taken into account as well. Figure 1 gives the number of wells on each island in parentheses after the island name. Table II gives the acronym designation of each well along with the island area and the depth of each cased and slotted well. Figure 2 shows the location of the well sites and surface pits cased only to 1 m depth (BDF designations) on Janet, the only multi-well island with a substantial freshwater inventory. Figure 3 shows the relative position of seven of eight wells on Yvonne, the island with the largest plutonium inventory at the Atoll. The wells on Sally are located along an axis across the middle of the island from the ocean to lagoon; XAR2 and XAR2A are near the ocean reef, XAR1 is approximately 20 m from the lagoon beach, and XAR3 is midway between XAR1 and XAR2A. The wells on the single-well islands of David, Ursula, Belle, Elmer, and Leroy are approximately at the geographical centers of the islands or in areas of planned resettlement. The wells were allowed to stabilize up to four months before sampling.

The first field sampling was in May 1974 followed by sampling trips during July-August 1974, January-February 1975 and October 1975. Our activities on each of these trips involved probing the wells to measure profiles of salinity, temperature, and dissolved oxygen, and water-sampling. Interim trips were made in November 1974, April-May 1975 and July 1975 to reprobe the wells and to maintain tide and rain gauges,

TABLE II. Island Wells: Summary.

Island Area (km ²)	Area (km ²)	Well I.D.	Depth of Slotted Casing below Ground Surface (m)
Janet	1.1	XEN1	89.5
		XEN2	70.3
		XEN3	19.8
		XEN4	28.4
		XEN5	64.4
		XEN6A	31.2
		XEN7	36.6
		XEN8	54.8
		XEN9	77.0
		AEN1	11.5
		AEN2	11.9
		AEN3	11.6
		AEN5	11.6
		AEN6	11.6
Yvonne	0.4	XRU1	53.0
		XRU2A	50.0
		XRU3	73.0
		XRU5	52.0
		XRU6	47.0
		ARU1	10.0
		ARu2	11.0
		ARU3	10.0
Sally	0.3	XAR1	65.0
		XAR2	34.0
		XAR2A	44.0
		XAR3	75.0
Leroy	0.06	ARI-1	11.0
		XRI-1	18.0
Ursula	0.11	ARO-1	10.5
Belle	0.19	ABU-1	11.0
Elmer	0.74	APA-1	12.0
David	0.44	AJA-1	11.0
Edna	0.007	XSA-2	68.0

A complete description of the analytical techniques used to measure all the chemical constituents in this program is beyond the scope of this report. Table III lists the measurements routinely made at the wells and on the groundwater samples. Since ours is the first extensive study of its kind at the Atoll, our measurements and techniques as they pertain to **groundwater** plutonium will be discussed in detail, but for the most part, **most** of the other techniques and results will be reported in forthcoming publications.

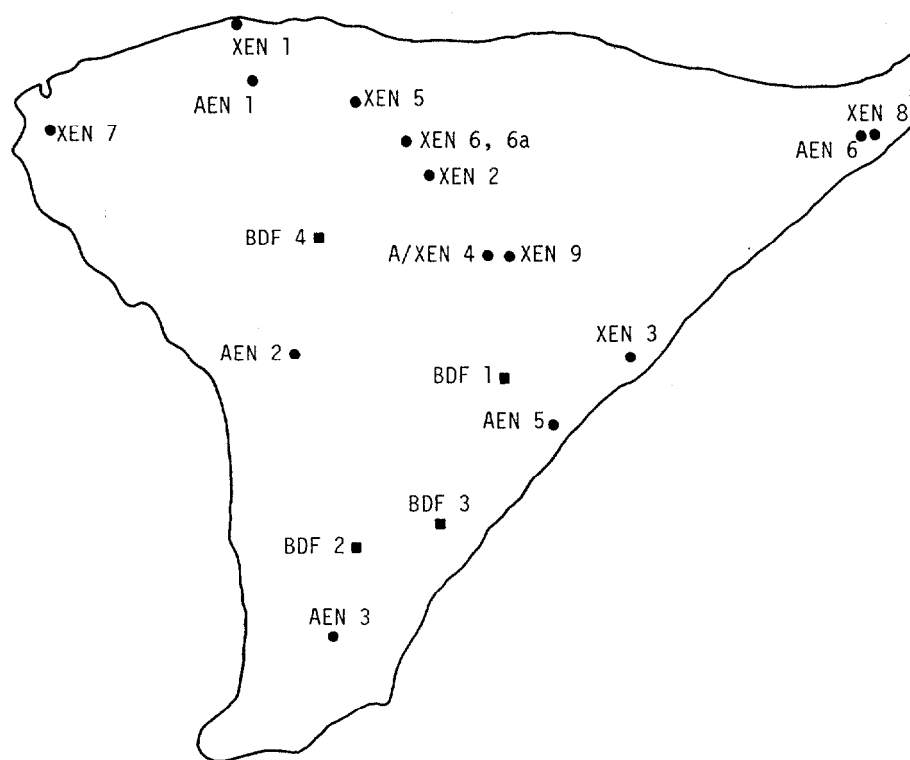


FIG. 2. Well locations on Janet.

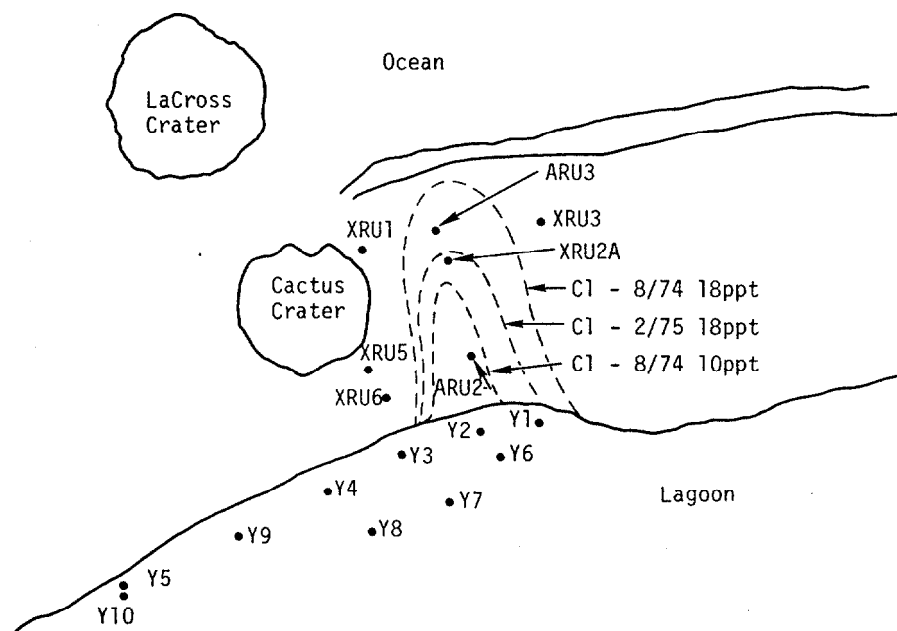


FIG. 3. Well locations on Yvonne. Also shown are the near-shore lagoon stations sampled in January 1975 (see text) and the surface chlorinity contours (18 ppt for August 1974 and February 1975 and 10 ppt for August 1974).

TABLE III. Measurements Included in Ground Water Program.

Field Measurements ^a			
Salinity profiles at each well		Rainfall measurements	
Temperature profiles		Soil leaching experiments	
Dissolved oxygen profiles		Pumping tests	
Tidal response (gauges) at specific wells		Lagoon and reef sampling	
		Ground water sampling	
Measurements on Selected Water Samples ^b			
pH	(1)	Strontium	(1)
Alkalinity	(1)	Chloride	(1)
Silicate	(1)	Sulfide	(1)
Phosphate	(1)	Sulfate	(1)
Ammonia	(1)	Coliform colonies	(1)
Nitrate	(1)	⁹⁰ Sr	(2)
Potassium	(1)	¹³⁷ Cs	(2)
Calcium	(1)	^{239,240} Pu, ²³⁸ Pu	(2)
Sodium	(1)	Gamma emitters detected by	(1,2)
Magnesium	(1)	spectrometry	
		³ H	(1,2)
		Particulate material:	(2)
		quantity and radionuclides	

^aCooperative effort of LLL and University of Hawaii personnel

^b(1) Analyses performed at University of Hawaii. (2) Analyses performed at Lawrence Livermore Laboratory.

A battery-operated pump draws groundwater from a predetermined depth through Tygon tubing to the ground surface. A variable-speed control permits pumping rates from a few millilitres per minute to 8 l/min. Either filtered or unfiltered water can be collected by valving off appropriate flow channels of the pump array. A different hose is used on each island and the entire pumping system is adequately purged at each well before sampling to minimize contamination. Depending on the island and well, a series of samples ranging in volume from 25 ml to 100 l is withdrawn for analysis of radionuclides, major and minor elements, nutrients, bacteria, and suspended material. Each sample container is rinsed at least three times with the sample water before collection is started. Except for the water collected for suspended material samples and coliform bacteria, all water is first filtered through a 0.4- μ m Nuclepore filter. If a heavy load of suspended material is anticipated or encountered, a 1- μ m filter cartridge is placed ahead of the 0.4- μ m filter. The filters are collected and subsequently analyzed for radionuclide content. The conductivity of the water is monitored continuously with an in-line meter to insure the collection of a sample of consistent chemical composition. We encounter, over very small depth intervals, variations in salinity ranging from a few percent to 100 percent that of seawater,

(seawater salinity averages, 34.5‰ range 34.3 - 34.6) and plutonium concentrations ranging from a fraction of a femtocurie per litre to hundreds of femtocuries per litre. Every effort is made to avoid contaminating the samples during field operations; we recognize, however that with the soil burdens in this contaminated environment, added to human and physical factors, the risk of sample contamination is high. Therefore, we maintain a rigorous quality-control program which involves the collection and analysis of duplicate samples, the preparation of blanks in the field, and the collection and sequential analysis of north equatorial surface seawater having $^{239,240}\text{Pu}$ levels of about 0.3 fCi/l. We also participate in national and international calibration exercises for the analysis of plutonium in selected water samples. The internal consistency of our data-- as shown by the blanks, the duplicates, and the extremely low-level oceanic samples-- indicates that sample contamination, even though it exists in the most rigorously controlled program, is minimal in our analyses. When suspect data have surfaced and subsequent sampling or duplicate analysis confirmed our suspicion of contamination, the suspect data were rejected.

One separation technique is used to isolate ^{90}Sr , ^{137}Cs , and plutonium radionuclides from all samples regardless of salinity. The filtered water sample (4 to 100 l) is acidified to pH 1 in the field and returned to the Laboratory for processing. Standardized mixed carrier solutions (containing stable carriers of Cs, Sr, Co, Bi, Sb, rare earths, and either ^{242}Pu , or ^{236}Pu and ^{243}Am) are added to the acidified sample which is then equilibrated by stirring for 6 h. Plutonium and other radionuclides are separated from the water and suspended samples using basically a published procedure⁹ with some modifications. The purified plutonium fractions are plated on platinum disks and counted on alpha spectrometers.

3. Results

Analyses of $^{239,240}\text{Pu}$ in the groundwater samples are presented in Tables IV through VI. The concentrations in filtered surface water are given in Table IV and those in filtered subsurface water are given in Table V. Table VI lists the data available for $^{239,240}\text{Pu}$ concentrations associated with the suspended material collected on 1- μm and 0.4- μm filters. Table VI also lists for each sample the logarithm of the distribution coefficient and the ratio of particulate plutonium to total plutonium. The distribution coefficient is the dimensionless ratio of the quantity of plutonium per gram of suspended material (dry weight) to that per gram of water. A considerable number of our suspended samples from July-August 1974 and January 1975 are still unanalyzed. None of the samples collected during October 1975 have been analyzed. The ^{238}Pu levels relative to those for $^{239,240}\text{Pu}$ are shown in Tables IV to VI for Yvonne. For most of the other samples the ^{238}Pu levels were associated with large errors or were below detection limits. Our detection limits for ^{238}Pu as well as for $^{239,240}\text{Pu}$ differed for each sample and depended on the chemical recovery, counting time, sample size, specific detector efficiency, and background. In some instances the upper limit of the isotopic ratio is shown.

Table VII summarizes the $^{239,240}\text{Pu}$ results detailed in Tables IV and V. The mean concentration and range for each island is computed for the period May 1974 to January 1975 for surface samples, samples collected at depths between the surface and 15 m, and samples collected at depths greater than 15 m.

TABLE IV. Plutonium Concentrations in Surface Groundwater Samples (Filtered Through 0.4- μ m Filter) during Month Indicated.

Island	Well	fCi/l $^{239,240}\text{Pu}$ (% σ) ^a			$^{238}\text{Pu}/^{239,240}\text{Pu}$ (% σ)		
		5/74	7-8/74	1-2/75	5/74	7-8/74	1-2/75
Janet	XEN1	6.0(10)	Lost	6.7 (3)	0.08(35)	Lost	0.03 (36)
	XEN2			2.0 (19)			
	XEN4	6.6(50)	12 (50)	7.8 (7)			
	XEN5			15.0 (9)			
	XEN6A			5.5 (16)			
	XEN9	6.0(60)		6.0 (60)			
	AEN1	12.0(40)		5.6 (9)	<0.3		<0.1
	AEN2	2.3(17)	3.2 (11) b 2.4 (16) b 2.6 (17) b	1.7 (16)	0.15(50)	0.15(42)	
	AEN3	3.3(38)	<2				
	AEN5	<3	<7	1.1 (56)			
	AEN6			7.0 (12)			0.25 (60)
	BDF1			3.8 (18)			
	BDF2			5.9 (11)			0.17 (70)
	BDF3			16.6 (6)			0.08 (60)
	XRU1			141 (2)			0.48 (4)
	XRU2A		Lost	54 (3)			0.54 (5)
	XRU3			6.6 (13)			
Yvonne	XRU5		6 (100)	175 (3)			0.47 (4)
	XRU6			69 (5)			0.45 (8)
	ARU1		45 (60)	31 (3)			0.063(13)
	ARU2		56 (40)	33 (6)			0.41 (8)
	ARU3			280 (6)			0.59 (3)
	XAR1			1.3 (70)			
	XAR2			12.3 (10)			
Sally	XAR2A			4.3 (21)			
	XAR3			3.8 (24)			0.32 (70)
	c4			1.4 (11)			0.48 (24)
	C7			1.8 (10)			0.01 (100)
	ARI1			5.9 (8)			0.26 (40)
Leroy	ARO1		2.5 (34)	3.3 (24)		co.11	
Ursula	ABM1	92 (4)		64.4 (3)	0.08(9)		0.04 (30)
Belle	APA1	<3	0.2 (100) b 0.22(75) b 0.2 (100) b	0.2 (15)			
Elmer			----	b			
			0.3 (100) b				
			0.2 (60) b				
David	AJA1	<3		0.32(25)			
Edna				27 (8)			
	XSA2						

^a(% σ) values in parentheses are 1 sigma counting errors expressed as percentages of listed values.

^bSamples taken during the well-pumping test on a single day. Concentrations are, respectively, at beginning, middle, and end of test.

TABLE V. Plutonium Concentrations: Subsurface Ground-water Samples during Month Indicated (Water Filtered through 0.4- μ m Filters).

Island	Well	Depth ^a (m)	fCi/l ^{239,240} Pu (%σ)			²³⁸ Pu/ ^{239,240} Pu (%σ)		
			5/74	7-8/74	1-2/75	5/74	7-8/74	1-2/75
Janet	XEN1	7	1.3(60) < 7					
		10			4.2(9)			0.06 (70)
		15	1.6(40) <15					
		20			3.6(9)		co.02	
		40			6.0(6)		0.04 (60)	
	XEN4	80			3.0(16)			
		12			5.8(8)			
	XEN9	25			6.0(5)			0.01 (100)
		25			2.8(24)			
	AEN1	7	<3					
		11			4.7(13)			0.08 (100)
	AEN2	10	5 (20)		2.5(17)			0.25 (100)
	AEN3	7	1.7(47)					
		10	1.3(50) 13 (60)		2.9(29)			
	AEN5	7	<3					
10		0.7(20)		2.6(18)				
Yvonne	XRU1	15			106 (4)			0.49 (4)
		40			287 (5)			0.53 (2)
	XRU2A	15			73 (9)			0.51 (7)
		35			663 (5)			0.57 (2)
	XRU3	8		6 (100)				
		15			34.2(5)			0.03 (84)
		45			9.9(8)			0.37 (13)
	XRU5	15			35 (7)			0.35 (15)
		45			126 (5)			0.45 (6)
	XRU6	15			168 (3)			0.47 (5)
		35			635 (4)			0.064(6)
	ARU1	9		38 (45)	123 (1)			0.15 (3)
	ARU2	10		27 (30)	87 (7)			0.43 (10)
	ARU3	10			240 (1)			0.54 (4)
	Leroy	ARI1	10			7.3(16)		
Ursula	ARO1	10		6.5(20)	16 (15)			0.01 (100)
Belle	ABM1	9		56 (18)	28.3(4)	0.14(70)	0.11 (20)	
Elmer	APA1	10	<3		3.2(12)			
David	AJA1	10	<3		1.1(18)			
Sally	XAR1	14			1.4(13)			0.09 (100)
	XAR2	20			19 (5)			0.03 (60)
	XAR2A	30			15 (10)			0.35 (20)
	XAR3	10			2.6(12)			0.05 (50)
		45			4.6(7)			0.09 (50)

^aDepths given are below ground level; water surface is typically 2.5 - 3.5 m below ground.

TABLE VI. Concentrations of $^{239,240}\text{Pu}$: Ground-Water Particulates (May 1974).

Island	Well	Depth (m)	Particulate (mg/l)	$^{239,240}\text{Pu}$ (% σ)		ln Kd ^d	Fraction in Particulates ^e
				(fCi/l) ^a	(fCi/mg)		
Janet	XEN2	Surf	10.4	21.0 (g) ^b			
				3.79(14) ^c	2.38	13.85	0.92
	XEN4	Surf	2.5	2.9 (30)	1.16	12.08	0.31
	XEN1	Surf	15.0	47.0 (9) ^b			
				0.95(13) ^c	3.20	13.19	0.87
		7	7.8	6.0 (18) ^c			
				0.26(33) ^b	0.80	13.33	0.82
	AEN1	7	7.5	1.5 (16)	0.2		
		10.8	26.3	1.3 (50)	0.05		
	AEN3	Surf	5.1	1.3 (60)	0.25	11.23	0.28
		7	3.1	0.9 (40)	0.30	12.08	0.35
		10	13.8	2.1 (25)	0.15	11.66	0.62
	AEN5	Surf	30.2	1.8 (90)	0.06		
		7	2.9	0.9 (70)	0.31		
	XEN9	Surf	0.8	2.4 (23)	3.0	13.12	0.29
Belle Elmer	AEN2	10	12.8	25.0 (7)	1.95	12.87	0.83
	ABM1	Surf	157	206 (2)	1.31	9.56	0.69
	APA1	Surf	4.4	0.2 (40) ^b			
				0.14(53)	0.08		
		10	19.6	0.4 (100) ^b			
				0.2 (30) ^c	0.03		

Mean ln Kd = 12.42 \pm 1.27Mean Kd = 2.5 $\times 10^5$ ^a0.4- and 1- μm filters were processed together except where indicated.^b1.0- μm filter only.^c0.4- μm filter only.^dCalculated by equation: $\ln Kd = \ln \frac{\text{fCi/mg particulate} \times 10^6}{\text{fCi/l water}}$ ^eFraction of $^{239,240}\text{Pu}$ in total particulates >0.4 μm .

To permit comparison of groundwater levels with soil concentrations, we list in Table VIII the log normal median soil concentrations and the ranges obtained from soil profile samples taken during the 1972-73 survey. The values for millicuries per square kilometer were computed to the depth sampled at each location and all island values were averaged to obtain the median value for the island. The distribution of plutonium on Yvonne was so complex that the island mean was obtained by an areal assessment of the soil burdens rather than from an arithmetic median. A mean soil density of 1.5 g/cm³ was used to convert activity per unit weight in a depth interval to activity per unit volume and area. From the ranges in soil values on all the islands, it is apparent that the distribution of plutonium is spatially heterogeneous,

TABLE VII. Mean $^{239,240}\text{Pu}$ Concentrations in Ground Water at the Depth Indicated, for the Period from May 1974 to February 1975.

239,240Pu (fCi/l ± S.D. from Mean)									
Island	No. Wells Avgd.	Surface	Range	No. Wells Avgd.	0 ≤ 15 m	Range	No. Wells Avgd.	≥ 15 m	Range
Janet	14	6.7 ± 4.5	1.1- 17	6	3.2 ± 1.7	0.7- 13	3	4.3 ± 1.6	3 - 6
Yvonne	8	101 ± 91	6 -280	8	97 ± 73	6 -240	5	344 ± 245	10 -635
Sally	6	4.2 ± 4.2	1.4- 12.3	2	2.0 ± 0.8	1.4- 2.6	3	13 ± 7	4.6- 19
Leroy	1	6.0			7.3				
Ursula	1	2.9 ± 0.4	2.5- 3.3		11.2 ± 6.7	6.5- 18			
Belle	1	77 ± 22	61 - 92		42 ± 20	27 - 58			
Elmer	1	0.7 ± 0.9	0.2- 2		3.2				
David	1	0.32 ± 0.06	0.2- 0.3		1.1				
Edna	1	27							

TABLE VIII. Log Normal Median Concentrations of $^{239,240}\text{Pu}$ in Soil Computed from Data in Ref. 1.

Island	Mean Core No, Length (cm)	of Core Samples Averaged	Ln Normal Median Concentration to Depth Sampled (mCi/km^2) and Range	Island Inventory to Depth Sampled (Ci)
Janet	150	13	1777 (318- 12902)	1.45
Sally	115	10	1244 (136- 3400)	0.57
Elmer	80	8	57.5 (13- 1262)	0.04
David	115	7	21.6 (3- 73)	0.01
Ursula	35	3	448 (140- 1000)	0.05
Leroy	35	3	459 (360- 540)	0.03
Edna	35	2	12422 (12260- 12580)	0.09
Belle	35	5	6168 (1627- 10297)	1.17
Yvonne	125	50	11678 (800-640000)	4.67
Yvonne Computed from areal assessment				26120
Yvonne N. Yvonne around 7 well sites				14340
				10.4

and because only a few profiles were averaged on many of the islands, the median values may not be truly representative of the total island burden. Often the shape of the vertical profiles indicated that appreciable quantities of plutonium were associated with the soil column below the maximum depth sampled. For these reasons, the values in Table VIII should be considered as reasonable estimates rather than absolute values of the average $^{239,240}\text{Pu}$ soil inventories.

World-wide fall out $^{239,240}\text{Pu}$ deposited to the 10-20° latitude band was $0.24 \pm 0.10 \text{ mCi}/\text{km}^2$ at the end of 1970 [10]. A cursory examination of more recent fallout data [11] indicates that only a small quantity has been added to this inventory since 1970. All the islands of Enewetak listed in Table VIII contain $^{239,240}\text{Pu}$ soil burdens from close-in deposition at levels many times greater than the fallout concentrations.

Significant fractionation of the plutonium isotopes in different regions over the Atoll is shown not only by the complex distributions of $^{239,240}\text{Pu}$ in the soils but also by the ratios $^{238}\text{Pu}/^{239,240}\text{Pu}$ in the groundwater (Tables IV and V). Hence, superimposed on the heterogeneous distribution of $^{239,240}\text{Pu}$ in the island soils is yet another heterogeneous distribution, that of ^{238}Pu , which indicates that the deposition of plutonium isotopes over the Atoll was not constant. Some of the plutonium inventory on any island may therefore be in different chemical or physical form and the environmental behavior of ^{238}Pu could be very different from that of $^{239,240}\text{Pu}$.

TABLE IX. Selected Concentrations of $^{239,240}\text{Pu}$ in Fresh and Marine Water

Location	Concn. Value (fCi/l)	Year	Ref. No.
Lake Superior (USA)	0.63	1973	12
Lake Michigan (USA)	0.73	1973	12
Lake Huron (USA)	0.63	1973	
Lake Erie (USA)	0.17	1973	12
Lake Ontario (USA)	0.25	1973	13
	0.72	1971	13
	0.41	1972	13
	0.27	1973	13
Tap water, Broomfield, Colorado (USA)	17.1	1970	14
Sewer treatment plant effluent, Livermore, California (USA)	7.9	1974	15
Mean surface concentration			
Enewetak lagoon	39.0	1972	16
Bikini lagoon	49.0	1972	16

4. Discussion

4.1. Water quality summary and plutonium surface concentrations

The islands David, Elmer, and Janet were found to support permanent freshwater lenses of different dimensions. The depth and the area occupied by the fresh surface water changes dynamically during the year in response to recharging from rainfall and vertical and horizontal mixing. Relative to seawater, the groundwaters show low pH, high alkalinity, reduced oxygen concentration, and high nutrient levels. Radiologically, chemically, and bacteriologically, the upper freshwater layers at David and Elmer appear usable for agriculture or household purposes if not further contaminated by human activities. The water could be consumed by humans with no serious ill effects, although by some western standards it would be minimally distasteful. The concentrations of $^{239,240}\text{Pu}$ in surface groundwater at David and Elmer were very low, comparable to those in the Great Lakes of the United States (Table IX).

No filtered surface groundwater on even the most contaminated islands contained plutonium at levels exceeding 0.3 pCi/l; however, unfiltered water probably will be used in household or agricultural applications. The concentration for **particulates** (average of those in Table VI) is approximately five times the concentration in filtered water. When particulate concentrations are added to those for filtered water, the total ~~surface-water~~ $^{239,240}\text{Pu}$ levels could range as high as 3 pCi/l in the most contaminated well sites.

Except for Northern Yvonne and Belle, the plutonium concentrations in the surface groundwater on the remaining islands sampled are less than the 1972 mean concentrations in the lagoons at both Bikini and Enewetak Atolls. The $^{239,240}\text{Pu}$ concentrations in most of the groundwater samples are lower than the 17 fCi/l reported in 1970 tap water from Broomfield, Colorado (Table IX). The former comparison indicates that groundwater plutonium is of little, but not necessarily zero, importance in maintaining the concentrations in the lagoons.

The maximum total $^{239,240}\text{Pu}$ concentrations found in surface groundwaters at Enewetak Atoll, including the particulate fraction, are currently less than 0.0% of the concentration guide for drinking water [17]. The present health hazard associated with ingesting the water from any location on Enewetak, at the present concentration of $^{239,240}\text{Pu}$, is less than the potential hazard from other pollutants in the water. For example, many locations on Janet have chemically acceptable groundwater with low plutonium levels, but the ^{90}Sr concentrations generally exceed the limit (10 pCi/l) set by the U.S. Public Health Service [18]. At other locations, including Sally and Yvonne and parts of Janet, the groundwater shows a seasonal transient fresher layer established during the rainy season, but the ^{137}Cs and ^{90}Sr concentrations in the surface waters at Yvonne and Janet would restrict even temporary usage. On all other islands sampled including Belle, Edna, Ursula, and Leroy, the chemical and/or radiological quality of the surface groundwater was poor throughout the year. It does not appear that a permanent freshwater lens can be maintained on Enewetak islands less than 0.2 km² in area.

Several small surface craters, averaging 10 m in diameter and 1 to 3 m in depth, were formed on Sally by high explosive nonnuclear tests in the early 1970s. These craters now have inventories of several hundred cubic meters of fresh water. The $^{239,240}\text{Pu}$ concentrations in two of these open craters (C₄ and C₇) are very low (Table IV) but show, interestingly, quite different $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios. One must conclude, on the basis of the different ratios, that the plutonium in these open craters was not derived from global fallout but from unique sources on the island. A complete assessment of water quality in these craters awaits the completion of other analyses.

During May and July 1974 large quantities of water were pumped from several wells, simulating use of the water by withdrawal over a one-day period, to follow the chemical and radiological changes in water quality in the surface-water layer as a result of continuous pumping for several hours at a constant rate. Secondly, since the total volume of the wells to the depth drilled was roughly 70 l (81 cm² cross sectional area and 8.5 m in depth), the tests would demonstrate whether the radionuclides were moving with the water or were simply residual activities in the hole, introduced during drilling or other physical operations at the well sites. On 17 May 1974, 5000 l were pumped from AEN2 on Janet. The inlet hose was located near the bottom of the well and the pumping rate was 34 l/min. Figure 4 shows the percentage seawater as a function of depth immediately before and after the test. The values for percentage seawater vs depth are computed from the in-situ conductivity measurements and compared to the conductivity of lagoon water after correcting for temperature. The surface and bottom concentrations in the well two days before the test were 3 ± 2 and 5 ± 3 fCi/l for $^{239,240}\text{Pu}$ and 56 and 57 pCi/l for ^{137}Cs . Both radionuclides were uniformly distributed vertically in the groundwater. Surface samples collected after the pump test contained 2.3 ± 1.4 fCi/l $^{239,240}\text{Pu}$ and 47 pCi/l ^{137}Cs . The water column was substantially disturbed as Figure 4 demonstrates. There was considerable

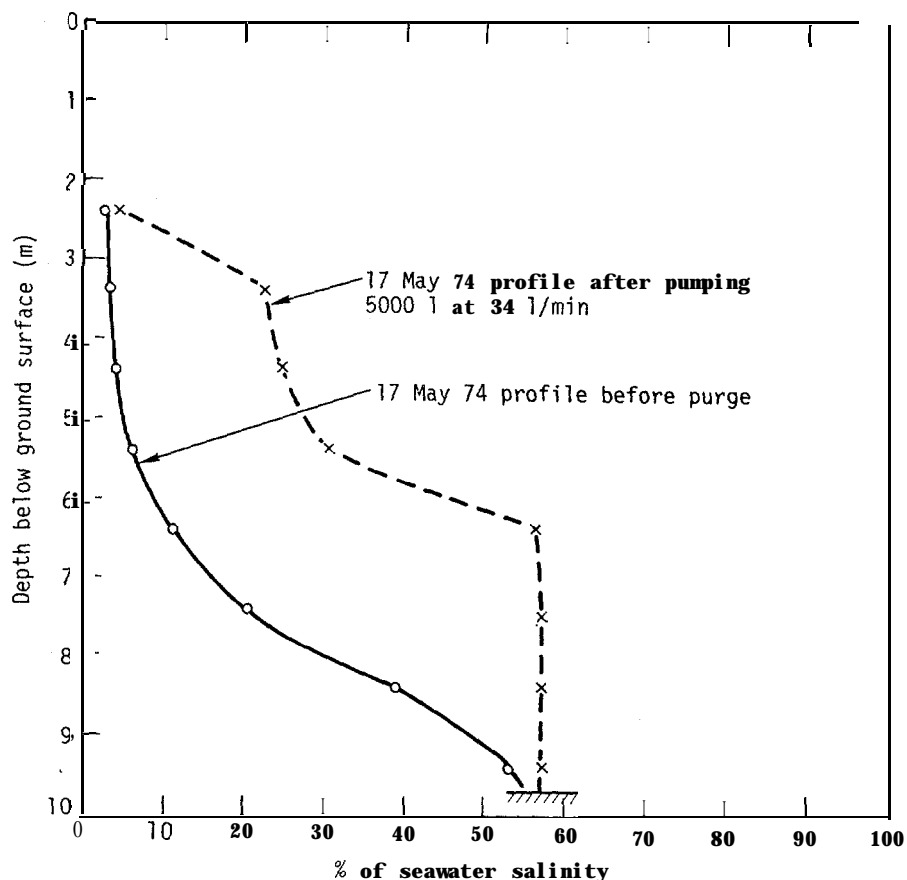


FIG. 4. Salinity profiles in AEN2 before and after the pumping test of 17 May 1974.

vertical intrusion of brackish water into the fresher surface layer although the actual surface salinity was little affected by the mixing. Surface ^{137}Cs levels decreased slightly, responding to the reduction in the amount of fresh surface water after the test, but the plutonium level was indistinguishable from the earlier value.

During July 1974, 800 gallons was pumped from AEN2 on Janet, APA1 on Elmer, and AJA1 on David. This time, 15-gallon surface samples were taken at the start, midpoint, and end of the test, and the conductivity was monitored throughout the entire experiment. Table X gives the analytical results of these experiments and Fig. 5 shows the structure of the water column before and after pumping at AEN2 and AJA1 and the in-line conductivity changes at AEN2 over the day. At AEN2 the conductivity changed significantly during the day. These changes were unaccompanied by such significant changes in $^{239,240}\text{Pu}$ and ^{137}Cs levels, although the ^{137}Cs concentrations did increase in the mid-day sample in response to the intrusion of fresher water in the surface layer. The entire water column again broke down; there was substantial vertical mixing of more saline water across the fresh-salt interface. At AJA1 and APA1, the recharging of the chemical and radiochemical constituents in the surface water was by horizontal intrusion of fresh groundwater into the well zone. The surface levels of $^{239,240}\text{Pu}$ at the three sites were, within our experimental accuracy, unchanged over the day. The "dissolved" surface $^{239,240}\text{Pu}$ in

TABLE X. Results from Day-Long Pumping Tests.

Well	Volume Pumped before Sampling (l)	$^{239,240}\text{Pu}$ (fCi/l) (% σ)	^{137}Cs (pCi/l)	^{90}Sr (pCi/l)	Cl (ppm)	Ca^{++} (ppm)
AEN2	0	3.2 (11)	55	19	3240	136
	1900	2.4 (16)	63	18	390	78
	3800	2.6 (17)	55	15	2080	122
APA1	0	0.2 (100)	1.3	0.12	1070	82.2
	1900	0.22 (75)	1.2	0.03	1050	84.5
	3800	0.2 (100)	1.1	0.04	1290	84.0
AJA1	0	Lost	0.78	0.017	460	87
	1900	0.3 (100)	0.83	0.011	770	92.8
	3800	0.2 (60)	0.83	0.023	930	92.2

the water immediately surrounding each well appears to be in equilibrium with the surrounding soil but its concentration at any time is unrelated to the change in freshness of the surface water.

When the surface-water concentrations during 5/74, 7-8/74 and 1-2/75 are compared (Table IV) the data, **except** at Elmer, present two examples of longer-term temporal changes of $^{239,240}\text{Pu}$ concentrations changes. Either the surface levels remained unchanged within the range of our analytical error between May 1974 and February 1975 (see the data from Janet, Ursula, David and part of Yvonne, Table IV) or decreased slightly in value (see results for part of Yvonne and for Belle, Table IV). Only at Elmer did the surface levels increase; at this site there were no other changes as significant as the order-of-magnitude increase in the plutonium surface levels. The Elmer well (APA1) is, however, unusual in one other respect. During our interim trip in November 1974 and our sampling trip in January-February 1975, roots of unknown origin and still unknown identity were found growing through the slotted casing at the depth of the groundwater surface. (The roots were absent from the well zone during the previous sampling programs.) In January 1975, broken root fragments 0.3 to 0.6 m long were pumped out with the water and collected along with other particulate **material** on the **filters**. We cannot be certain whether or not the presence of the roots in the surface groundwater is responsible for elevating the plutonium surface concentrations but we are investigating this possibility. It is possible that the roots of fast-growing **phreatophyte** plants could cause mechanical reworking of upper soils to greater depths in the soil column, or that plutonium taken up by surface roots of some atoll plants is transferred internally to deeper root zones. The transferred plutonium in either decayed or live roots could be exchanged or transferred to the ground water. Such mechanisms could have an impact on the quality of the groundwater and on future rehabilitation.

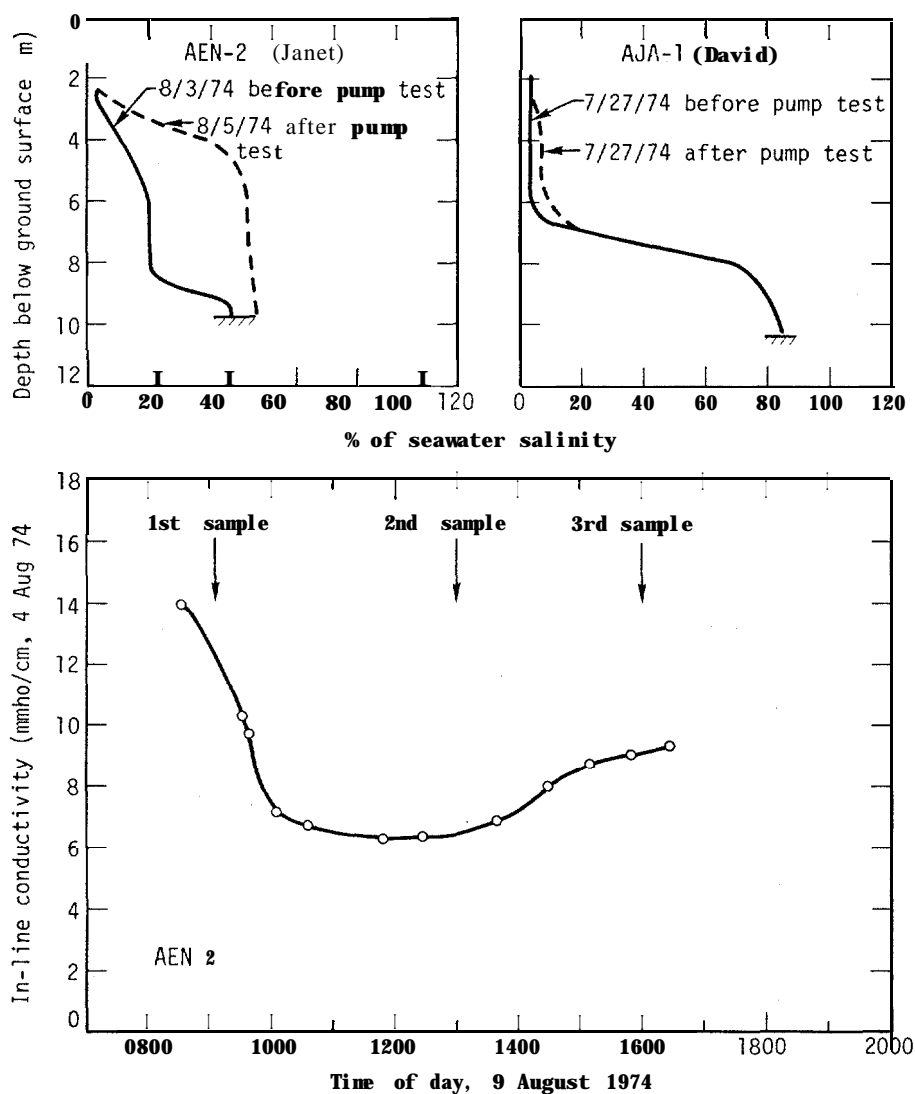


FIG. 5. Characteristics of water columns during pumping tests of July-August 1974. Upper left salinity profile at AEN2 (Janet) before and after test. Upper right, salinity profile at AJA1 (David) before and after test. Lower, in-line surface conductivity changes during pumping at AEN2 (Janet).

On Janet, at those wells sampled two or more times between May 1974 and February 1975, no significant temporal variations in $^{239,240}\text{Pu}$ concentrations in the surfacewater were evident. More significant changes, by comparison, are noted in the concentrations of ^{137}Cs , ^{90}Sr and the major elements. Table XI compares these differences at XEN1, XEN4, AEN2, and AEN1 (see Fig. 2 for locations). It appears that at all of these locations the processes regulating the levels of $^{239,240}\text{Pu}$ in the surface groundwater are independent of the processes controlling ^{137}Cs and ^{90}Sr distributions. Of the 13 surface samples collected at Janet in January-February 1975, three had chloride concentrations greater than 10 ppt (parts per thousand) four samples had surface chlorinities between 2 and 10 ppt and the remaining six wells had surface chlorinities less than 2 ppt. The mean surface $^{239,240}\text{Pu}$ concentrations in these respective chlorinity intervals were 5, 6.1 and 7.6 fCi/l. A slight dependence on

TABLE XI. Temporal Concentration Changes at Four Wells on Janet (Surface Water).

Well	Date Sampled	$^{239,240}\text{Pu}$ (fCi/l) (% σ)	^{137}Cs (pCi/l)	^{90}Sr (pCi/l)	Cl (ppm)	Ca^{++} (ppm)
XEN1	5/74	6.0 (10)	134	9.1	1720	93.5
	7/74		121	20.4	1500	88.0
	1/75	6.7 (3)	83		2800	52.0
XEN4	5/74	6.6 (50)	76	1.6	10170	236
	7/74	12.0 (50)	125	0.3	4450	128
	1/75	7.8 (7)	58		456	70
AEN2	5/74	2.3 (17)	47	26	2730	190
	7/74	3.2 (11)	55	19.1	3240	136
	1/75	1.7 (16)			2340	93
AEN1	5/74	12 (40)	118	0.31	3110	132
	1/75	5.6 (9)	87		1770	93
		$\bar{X} = 90$ $\bar{X} = //$ $\sigma = 32$ $\sigma = //$				

water freshness is indicated by these values, but, considering the large range in surface chlorinity and the small range in $^{239,240}\text{Pu}$ levels, the data rather suggest that plutonium surface concentration is independent of quantity of fresh water. Plutonium concentrations in surface groundwater were not significantly affected by the seven month temporal changes in rainfall supplying fresh water to the island reservoir and were unrelated to the standing quantity of fresh surface water in the reservoir.

The wells ARU3, XRU2A and ARU2 on Northern Yvonne (Fig. 3) are located in a slight depression between the southern rim of Cactus crater on the north and elevated ground to the south. The chloride, ^{137}Cs and, $^{239,240}\text{Pu}$ concentrations in surface groundwater for these and surrounding wells and for the local marine environment are tabulated in Table XII. Reference to the surface chlorinity contours in Fig. 3 and the data in Table XII shows that the surface groundwater in this depression is fresher than the surface water in the wells to the north and south. The area must function as a catchment basin for freshwater runoff around the crater. Its mean residence time is short, as can be estimated by comparing the surface areas occupied by the 18-ppt chlorinity contours during August 1974 and February 1975. During these periods the surface water was always freshest at ARU2 compared to XRU2A and ARU3. The water freshness increased along an axis across the island from the ocean to the lagoon indicating that the fresh groundwater flux in this region is preferentially lagoonward. Off-shore lagoon samples collected in January 1975 (see Fig. 3 for locations and Table XII for concentrations) show higher ^{137}Cs levels near the shore around the extension of the axis of presumed groundwater flux. The ^{137}Cs levels were much higher in the surface waters at locations Y1, Y4, and Y6 (approximately 30 m from shore) than at stations up to 100 m north or at stations 50 m closer to the lagoon. If these higher concentrations were regulated by sediment-leaching mechanisms, wave action, surface runoff, or other marine processes, much more uniform concentrations would be anticipated over the

TABLE XII. Surface $^{239,240}\text{Pu}$ Concentrations and Other Data at North Yvonne Well at Near Offshore Lagoon Stations (Shown in Fig. 3), and in Cactus Cr

	Cl^- (ppt)	^{137}Cs	$^{239,240}\text{Pu}$	$^{238}\text{Pu}/2$
	8/74	2/75	8/74	2/75
Northern wells				
XRUI		19.2		141
ARU3	13.0	19.3		280
XRUI	18.5	19.0	Lost	6.6
XRUI2A	11.0	17.7	Lost	154
XRUI5		19.4		175
XRUI6		18.7		69
ARUI2	8.8	16.5	56	33
Southern well				
ARUI	16.0	19.0	45	31
Cactus Crater				
Surface (high tide)				92
Bottom (high tide)				80
Yvonne ocean reef				
water (opposite ARUI)				97
Offshore lagoon				
stations				
Y1		1.63		178
Y2		2.14		
Y3		1.73		
Y4		2.40		
Y5		0.89		
Y6		3.15		
Y7		0.70		
Y8		0.53		
Y9		0.58		
Y10		0.55		
				0.18 (6)
				0.18 (10)
				0.17 (11)
				0.18 (2)
				0.17 (4)
				0.18 (2)
				0.20 (10)

entire area sampled. The near-shore lagoon data indicate that exchange of groundwater with the lagoon probably occurs only in specific regions of an island; other areas containing barriers, such as surface or buried beach rock, restrict the outflow of groundwater. High levels of $^{239,240}\text{Pu}$ were found at all lagoon stations sampled off Yvonne but plutonium, unlike ^{137}Cs , was highest in concentration north of stations Y1, 4, and 6; by comparison, concentrations were lower near the presumed outflow region. In addition, all surface lagoon water at these stations had $^{238}/^{239,240}$ isotope ratios of about 0.18, a value significantly different than the values of 0.4 to 0.5 for groundwater at ARU3 and ARU2. If the emergent ^{137}Cs concentration in the groundwater from Yvonne was 330 pCi/l (similar to the groundwater surface level at ARU2) in January 1975 by the time the water reached the position of Y1 and Y6, the ^{137}Cs had been diluted at least 10- to 20-fold by lagoon water having a mean concentration of 0.3 pCi/l. A similar reduction in the $^{239,240}\text{Pu}$ levels in the emergent groundwater mixed rapidly with the higher-concentration water from the north reef, would make the groundwater plutonium flux and the isotopic ratio indistinguishable in the lagoon. It is significant that the reef, a few hundred meters north of the wells, is a source contributing $^{239,240}\text{Pu}$ to the lagoon. The small study area around the crater illustrates that many mechanisms contribute to the aquatic recycling of plutonium radionuclides in this atoll environment.

Returning to the results in Table XII, it is apparent that during both sampling periods, the ^{137}Cs concentration gradient paralleled the freshwater gradient. The quantity and rate of ^{137}Cs recharging the groundwater are directly related to the freshwater flux and quantity. The concentration gradient of $^{239,240}\text{Pu}$ in the surface layer during January 1975 was exactly opposite to that of the freshwater and ^{137}Cs . The concentration was highest at ARC3 and decreased along the island axis to ARU2. The $^{238}/^{239,240}$ ratio in the surface water decreased toward the lagoon. The change in ratio could be associated with some unknown fractionation phenomena occurring during horizontal migration, but it is more likely that either (1) the reduction is merely the result of mixing water of lower specific activity with water of lower $^{238}/^{239,240}$ ratio from the south (see XRU3 values, Table XII) or (2) simply that the $^{238}/^{239,240}$ values in the soils and groundwater in this region are very heterogeneous. These data demonstrate once again that the processes affecting the lateral mixing of $^{239,240}\text{Pu}$ differ significantly from those controlling ^{137}Cs . The flux of plutonium is probably impeded and controlled during transport by exchange and/or sorption processes within the coral matrix of the soil.

The presence of detectable quantities of $^{239,240}\text{Pu}$ in the surface groundwater on all islands sampled is clear evidence that, regardless of the mechanism involved, small quantities of plutonium radionuclides have migrated to the groundwater reservoirs since the middle to late 1950s. Interestingly, the median soil burdens of $^{239,240}\text{Pu}$ for the islands (Table VIII) relate directly to the respective surface groundwater mean concentrations in Table VII. Figure 6 shows the logarithm of the mean $^{239,240}\text{Pu}$ concentrations in surface groundwater (fCi/l) plotted against the logarithm of the median concentrations in soil (mCi/km²). A best fit linear regression analysis of the data, using the units indicated, yields the following relationship:

$$\frac{^{239} \text{ } ^{240} \text{ Pu (fCi/l) in surface ground water}}{^{239} \text{ } ^{240} \text{ Pu (mCi/km}^2\text{) in the respective island soil}} = 8.0 \pm 4.9 \times 10^{-3} (1)$$

the value of k is $2 \pm 1 \times 10^{-5}$. On the average then, 0.002% of the plutonium inventory of any island is contained in the top 2.5 m of the groundwater. Worldwide fallout $^{239,240}\text{Pu}$ on "uncontaminated" Pacific atolls in the 10-20° latitude band may be taken, from our earlier estimate, as 0.3 mCi/km^2 . Equation 1 predicts that only 2 aCi/l of $^{239,240}\text{Pu}$ should be expected in the surface groundwater at these atolls. We are currently analyzing groundwater samples from Bikini Atoll to test the applicability of this model at other atolls. Reference to the next section shows, however, that $^{239,240}\text{Pu}$ is not restricted to the surface layers but is distributed to great depths in the groundwater column. Hence, the surface-layer inventory is only a small fraction of the total quantity distributed throughout the entire island reservoir. Estimation of groundwater inventories and of surface and deepwater mixing and flushing rates is critical if we are to determine plutonium residence times in the island soil column. We are not yet in a position adequately to discuss these parameters.

b. Vertical distributions and particulate concentrations

The $^{239,240}\text{Pu}$ is detected at depths as great as 80 m (XEN1 in Table V) in the groundwater reservoir and is detected at all intermediate depths sampled. Our pumping experiments, numerous other tests, and our other chemical and radionuclide data from the wells has convinced us that the occurrence of plutonium at depth in the groundwater reservoir is real and not a phenomenon related to possible physical disturbance or alterations to the reservoirs by drilling operations. These deep-water data indicate that plutonium is very mobile throughout water-saturated coral-soil environments.

Figure 7 shows the percentage concentrations of ^{137}Cs , fresh water, and $^{239,240}\text{Pu}$ vs depth in the groundwater column at XEN1 on Janet. There is a close similarity between the vertical distributions of fresh water and ^{137}Cs . Curve A was constructed assuming that ^{137}Cs behaves as a solute throughout the reservoir and that all activity variations at any depth in the water column are accountable for by the freshwater component of the vertically mixed fresh and ocean water. The ^{137}Cs activities associated with the freshwaters (A_{fw}) and saline waters (A_{ow}) can be computed from a least squares regression analysis of the XEN1 data by the following equation:

$$\text{Total activity } (A_t) = A_{fw} \left(\frac{\text{Cl}^- \text{ sample}}{\text{Cl}^- \text{ ocean}} \right) + A_{ow} \left(1 - \frac{\text{Cl}^- \text{ sample}}{\text{Cl}^- \text{ ocean}} \right), \quad (2)$$

where Cl^- is the chloride concentration and A_t is the measured ^{137}Cs concentration at the specified depth. The Cl^- concentrations in island rainwater and in freshwater samples collected by leaching upper layers of coral soil are insignificant. Therefore, we assume that any Cl^- concentration at depth in the water column is due only to the oceanic component of the water. The computed mean A_{fw} and A_{ow} values for ^{137}Cs in XEN1 are, respectively, 102.8 and 6.2 pCi/l.

Curve B for ^{137}Cs in Fig. 7 was constructed from equation 2, assuming that the Cl^- and ^{137}Cs concentrations are conservative at only the three deepest depths sampled. The computed means A_{fw} and A_{ow} for this curve are, respectively, 127.2 and 0.46 pCi/l. Curve B fits the deep-water data much better than curve A, but the extrapolated surface value is 40% greater than the measured surface concentration. The ocean reef water off XEN1 in January 1975 contained 0.38 pCi of ^{137}Cs per liter,

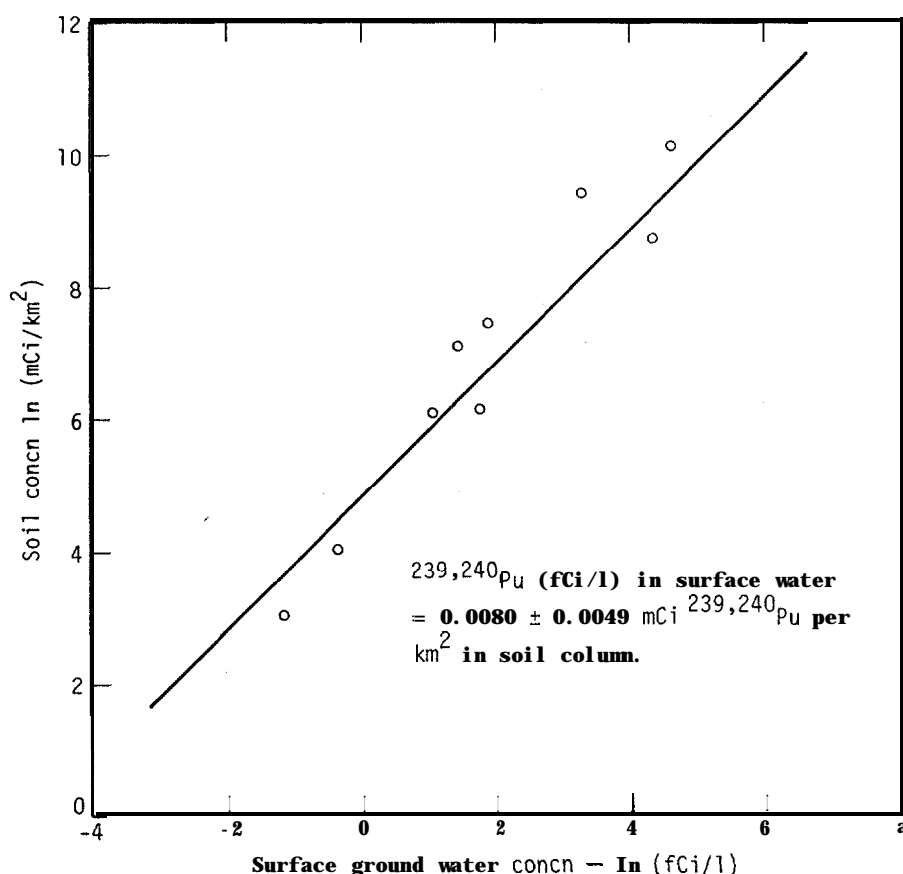


FIG.6. Soil/groundwater relationships for $^{239,240}\text{Pu}$: median soil concentrations from Table VIII versus mean surface groundwater concentrations from Table VII,

Considering the large spread in the soil and water levels and the fact that the mean concentrations from several islands are derived from only one or two measurements, the relationship is much better than one would anticipate. The significance of this relationship is, that regardless of island size, geographical location, groundwater residence time, or possible differences in the chemical or physical state of soil plutonium, the concentrations in surface groundwater are directly proportional to the measured soil burdens. This suggests that $^{239,240}\text{Pu}$ is readily exchanged between the island soil column and the recharging fresh rainwater. The quantities reaching the surface ground reservoirs, which for all practical purposes are at the same depth below ground surface, are regulated by the soil inventories, the quantity of freshwater recharging the surface layer, and the magnitude of the distribution coefficient for plutonium between water and the island soil components.

If the surface layer is arbitrarily defined as the distance to the point of maximum chlorinity inflection in the water column, the average thickness of this layer for all islands is 2.5 m. Converting equation (1) to read:

$$\frac{^{239,240}\text{Pu} \text{ (mCi/km}^2 \text{) in first 2.5 m of groundwater}}{^{239,240}\text{Pu} \text{ (mCi/km}^2 \text{) in island soil}} = k$$

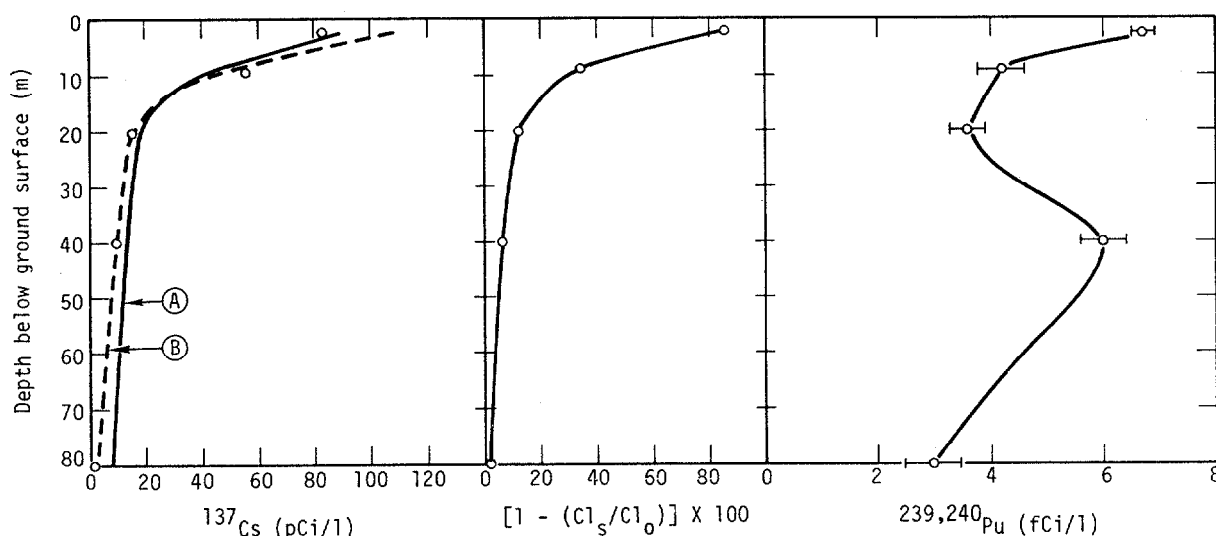


FIG. 7. Vertical profiles of ^{137}Cs , Cl^- , and $^{239,240}\text{Pu}$ at XEN1 (Janet). The percentage freshwater in the water column is $1 - (\text{Cl}_s/\text{Cl}_o) \times 100$, where Cl_s is the salinity of the sample and Cl_o is the salinity of the ocean.

which is in better agreement with the A_{ow} value of 0.46 pCi/l generated in the second case. The deviations of the measured ^{137}Cs concentrations from curve A are more than anticipated from the accuracy of our measurements. We believe that the data demonstrate that the constituents in the upper fresher groundwater layer and the deeper layers cannot be treated conservatively, because the mean horizontal velocity of the surface water and its dissolved chemical species must differ greatly from the vertical mixing velocities. The net effect of different surface and vertical velocities could explain the different surface ^{137}Cs concentrations predicted by curve B. The bottom concentrations show that there is a conservative relationship between the quantity of freshwater and ^{137}Cs , but the advective surface velocities and amounts of freshwater and ^{137}Cs recharging the surface layer are sufficient to change the $^{137}\text{Cs}/\text{Cl}^-$ ratio relative to the ratio in the deep reservoir at any time. The concentration profile of $^{239,240}\text{Pu}$ bears little resemblance either to the freshwater or the ^{137}Cs distributions. The processes that control the vertical distribution and flux of plutonium in the groundwater column are independent of the processes that control the vertical mixing of ^{137}Cs . The vertical distributions of plutonium at any one well are unrelated to the distributions found on other islands or for that matter at different wells on the same island. On Sally, for example, $^{239,240}\text{Pu}$ at XAR1 is uniformly distributed throughout the groundwater column to the depth sampled while at XAR2, XAR2A and XAR3 the concentrations increase with depth. On Yvonne, all the wells except ARU1 are located within an area of 5800 m². At ARU3 the surface level of $^{239,240}\text{Pu}$ is higher than the bottom concentration. At XRU1 and XRU5 the concentrations are minimal at about 15 m depth and at XRU3 they are maximal at the same depth. At the remaining sites the $^{239,240}\text{Pu}$ concentrations increase from surface to bottom. At XRU6 in January 1975 the 238/239,240 ratios for the surface and 15-m samples were the same, 0.46. For the 35-m sample from this well, the ratio was 0.064. Comparison of these ratios in the same well indicates that in addition to surface horizontal mixing and Vertical mixing, deep lateral groundwater fluxes move plutonium on some islands.

No other physical or chemical data have confirmed this proposal so far, but it does underscore the necessity for careful hydrologic modeling to estimate plutonium transport vectors and rates in the ground water.

In Table VI a mean K_d value of 2.5×10^5 for $^{239, 240}\text{Pu}$ was computed from the available concentration data from particulate and water samples. Seven soil samples containing high but different levels of plutonium were obtained from different horizons in Yvonne soil and equilibrated separately in the laboratory with uncontaminated sea water. Fractions of the water were removed at intervals, filtered through a $0.4\text{-}\mu\text{m}$ filter, and analyzed for plutonium. After equilibrium was reached, the mean K_d for these samples was 8×10^4 . The close similarity between our field and laboratory results suggests that the control of groundwater plutonium concentrations is maintained through exchange processes with small suspended fragments which migrate to the water reservoir or, again by exchange processes, with the dissolved ($< 0.4\text{-}\mu\text{m}$) plutonium transported with the recharging fresh water, and with the water-saturated soil. The concentration of plutonium at any depth or location appears to be regulated by the quantity and specific activity of the labeled particles in the water column.

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DISCUSSION

E.P. HARDY: Have you measured, or do you intend to measure, any of the transuranics of higher mass — such as americium and curium — in the groundwater samples ?

V.E. NOSHKIN: We do intend to measure americium, but we have been kept busy with plutonium measurements so far.

H. F. MacDONALD: You mentioned that one of the motives for this study was to investigate the suitability of the larger islands in the Enewetak Atoll for future habitation. In this sense, your results would be relevant to the reoccupation of areas around nuclear installations following a major accident. It would therefore be interesting to know whether there are any additional data on levels of airborne plutonium and the resultant dose rates on the atoll. Is the area at present suitable for human occupation?

V.E. NOSHKIN: That question goes somewhat beyond the scope of our paper. I might refer you to the Enewetak Radiological Survey, cited as Ref. [1] here, which contains dose assessments for the different pathways by which radionuclides reach man — inhalation and the food-chain — as well as for external radiations in both the marine and the terrestrial environment. This is probably your best reference.